Responses to Reviewer #2

Thank you to both reviewers for their comments. We have continued to work to clarify the methods and discussion in the manuscript.

The paper presents a nice way to partially isolate the affects of cloud droplet distribution shape on condensation/evaporation by comparing condensation/evaporation rates in a bulk model to a bin model. Statistics are produced for average condensation/evaporation rate binned in terms of distribution properties such as average drop distribution diameter. It is shown that when comparing bulk to bin condensation/evaporation rates, removing the effect of of the distribution shape will generally produce better comparision, expect when evaporation mass fraction is high. While the method is useful more discussion on how to apply the method to improve bulk models is needed.

It is beyond the scope of this paper to determine how to improve the representation of the shape parameter in bulk schemes. The purpose of the paper is to show that the shape parameter is responsible for a large degree of the disagreement between bin and bulk schemes (in terms of condensation/evaporation), and to argue that more work is needed by the community to improve the representation of the shape parameter in models. We are not suggesting that the methods we have used to show that the shape parameter is important should be the same methods used to "fix" bulk schemes. This is now explicitly stated in the conclusions.

General Comments:

You discuss comparing process rates between bin and bulk in order to improve bulk schemes. So how do you use your analysis to improve bulk schemes? It seems like the parameter space of various values of number concentration, distribution shape, and average cloud droplet size is so large that this study, although useful, would have trouble providing direct improvement to bulk models? Is this correct or is there a good way to use bin models to inform bulk models for condensation/evaporation?

Yes, the reviewer is correct. It would be difficult to use our analysis methods to improve bulk schemes. Though not the topic of the paper, we do feel that searching for robust empirical relationships between the shape parameter (or relative dispersion) of simulated distributions and other cloud properties may be one way to use bin schemes to inform bulk schemes.

Why not use values from the curves from Fig.1 when choosing simulations to run?

This is certainly an approach we could have taken. However, the methods we used to choose values should not impact our results.

Line 95: You state "the lack of a prognosed shape parameter for the cloud droplet size distribution in the bulk scheme is often the primary source of difference between the two schemes." Why only used a fixed shape parameter in the bulk simulations? Why not diagnose a value? Would diagnosing a value provide better results? How would diagnosing a value change your results? Perhaps diagnosing the shape parameter would leave to better agreement between bulk and bin when evaporation fraction is high.

No good ways to diagnose the cloud droplet shape parameter exist to our knowledge. Some diagnostic equations do exist, but as we discuss in Igel and van den Heever 2017a, most do not seem appropriate for high resolution simulations such as these. We absolutely agree that diagnosing the shape parameter would lead to better agreement – that is essentially one of the points we are trying to make.

Specific Comments:

Abstract Line 16: I don't agree that the statistics are novel. Maybe replace with "statistically" We have made the change.

Manuscript

Line 35: The shape is sometimes fixed as well

We are not sure what the reviewer means by "shape". We state that "a function is assumed to describe the shape of the size distribution ..."

Line 44: Probably can remove discussion on ice as it it irrelevant to the paper It has been removed.

Line 64: What parameter are you talking about? Thank you, the parameter is the shape parameter, and this is now explicitly specified.

Line 107: Remove this line. We have removed it.

Line 109: PDF is already defined Thank you. We now just say PDF.

Equations 2 and 3 need periods at the end of the sentence

Thank you, they are now included.

Line 143: What do you mean by this? Do you have a larger range of bins and thus more statistics? This is a type. The word should be "wide", not "wider"

This is a typo. The word should be "wide", not "wider".

Line 146: How deep are the clouds in the simulations? A grid of 3.5km high seems far to shallow.

No cloud top exceeded 2.85km from the surface, and the vast majority of clouds had tops below 2.5km.

Line 196: extra comma

Thank you, it has been removed.

Line 219: Certainly clouds exist between 99-101% RH, and certainly signatures of the drop distribution properties should up between these RHs. Why not just include the analysis? Also, what percentage of the grid exists between these humidities?

Yes, clouds certainly do exist at RH of 99-101%. In the low aerosol simulations, they are 28-33% of the cloudy points, in the moderate aerosol simulations, they are 42-64% of the cloud points, and in the high aerosol simulations, they are 64-67% of the cloudy points. In the revised paper, we have included all data, and the results are very similar. We have also added a section about the impact of relative humidity on the comparison of the two schemes. This section more clearly demonstrates and explains why RH close to 100% leads to a worse comparison between the two schemes.

Line 234: Extra ")" Thank you, it has been removed.

Could you possible get rid of Table 2 and incorporate the standard deviation data into the figures?

Yes. We have removed Table 2 and listed the standard deviation data in the legends of each plot.

Line 249: Evaporation seems to tend towards bulk in Fig. 3A Agreed. This is now noted.

Line 251: Is the long tail in the condensation or evaporation? The evaporation.

Line 289: Briefly describe the method used to fit the bin distributions. We now include such a description. It is reproduced here:

Here we used the maximum-likelihood estimation (MLE) method. For our problem, the log-likelihood function (ln(L)) is defined as

 $\ln L = \frac{1}{N_t} \sum_{i=1}^{15} N_i \ln n(D_i)$ (4)

where $n(D_i)$ is the value of the gamma PDF (Eq. 1) for D_i with unknown values of the parameters D_n and v. The function is normalized by the total cloud droplet concentration N_t in order to remove N_t as a free parameter in Eq. 1. As indicated by its name, the MLE method seeks to maximize the log-likelihood function given by Eq. 4. To do so, we used the MATLAB function fmincon to find the parameter values that minimized -1^*L .

Line 301: What percentage of the data had best-fit shape parameters less than 1? Did it occur frequently?

For BIN100, BIN400, and BIN1600, it was 4.5%, 5.1%, and 8.6%, respectively. These values are now specified in the manuscript.

Correcting the data seems to make the orange line in Fig. 3C worse. Why?

This is a question that we have spent a considerable amount of time thinking about. We think that the reason may be that differences in the schemes caused by sub-time stepping are more important than the shape parameter differences in this case, but only because the shape parameter used by the BULK simulation was similar to the best-fit shape parameters in the BIN simulaiton.

Line 351: What does an NRMSE of 2 mean?

There is no particular meaning of an NRMSE of 2, except to say that it is worse than assuming a fit that is independent of the predictor (in this study diameter) and equal to the mean of the data. The NRMSE is defined here as

 $NRMSE = \frac{\sqrt{\sum_{i=1}^{15} (y_i - \hat{y}_i)^2}}{\sqrt{\sum_{i=1}^{15} (y_i - \bar{y})^2}}$

where y_i is the simulated probability density of droplet concentration in the ith bin, \hat{y}_i is the fitted probability density of droplet concentration in the ith bin, and \bar{y} is the average value of all y_i 's. The sum

runs from 1 to 15 since we have 15 bins containing cloud droplets. Thus if the fitted probability density \hat{y}_i is equal to \bar{y} for all i, the NRMSE is 1.

That said, we did find an error in our calculation of NRMSE. It has been corrected, and as a result, values greater than 1 are now extremely rare. The new cumulative distributions are shown below.



Figure 4. Needs to be explained better in the text. Is it showing that approximately 80% of the cloudy grid points have an NRSME<0.6? Why is 0.6 considered appropriate?

We agree that 0.6 was an arbitrary cutoff value. We have removed this figure and replaced it with a new figure in order to make the same arguments, but in a way that does not require the use of an arbitrary cutoff. Here we show the new Figure and its description.

To explore simultaneously the impact of NRMSE and fractional mass change on the comparison of bin and bulk scheme condensation and evaporation rates, we also calculated the mean NRMSE and fractional mass change of each of the joint *S*, *N*, and \overline{D} bins in addition to the corrected mean ln(ratio) for each bin that we have shown previously. In this analysis, we have excluded points with relative humidity between 99.5% and 100.5%. Joint bins with similar mean NRMSE and fractional mass change were grouped together to find a mean of the corrected mean ln(ratios). Joint bins from all simulation pairs were included. The results are shown in Figure 7, again for condensation and evaporation separately, where colors show the mean of the corrected mean ln(ratios) as a function of NRMSE and fractional mass change.



Line 395: It can account for some changing drop distribution properties, but not large changes to the shape parameter that may occur when evaporation rates are high. Is this large change in distribution shape during evaporation that the bin model can capture the reason for the difference in Fig. 5A?

Yes, we believe that the bin model's ability to account for the changing distribution during evaporation is the reason for the large differences in Figure 5a (which has been removed since it used an arbitrary cutoff) and the new Figure 7a (shown above).

Maybe move Fig. 4C and 4D to their own figure?

Thanks for the suggestion. With the slightly new development of the discussion, we felt it was better to leave them all as one figure.

Line 443: But what does that high evaporation fraction do to the bin distribution?

We are not entirely sure what the reviewer is asking. Like the reviewer mentioned above, the bin scheme is able to deal with large changes to the size distribution during evaporation in ways that the bulk scheme cannot. In part this is due to the way that the bin scheme uses sub time steps during evaporation and condensation. We believe that it is these differences in the schemes that lead to the dependence on the evaporation fraction.

Conclusion 1: What assumptions were made for the aerosol distribution in the bin model? Was it initially assumed to be a gamma distribution? If so, then it is no surprise that using a gamma distribution would be a good bulk assumption compared with bin.

No, the aerosol distribution was initially lognormal with a median radius of 40nm and a spectral width of 1.8. These details are now included in the description of the simulations. That said, we agree that it may not be surprising that using a gamma distribution is a good assumption. In the revised manuscript, this conclusion has been removed.

Conclusion 2: (Line 468) You didn't show that sub-time stepping is important. Remove this sentence.

The reviewer is correct, we did not directly show that the sub-time stepping is important. However, we did show that the fraction of mass evaporated is important, and we believe that the reason for this

importance relates to the sub-time stepping used by the bin scheme. In the revised conclusions, we do not mention the sub-time stepping.

Line 473: Specify the conditions that this applies for: Low evaporation fraction, humidities >101%, etc.

Thank you for the suggestion. We have modified the statement to specify that the conclusion only applies to situations when relative humidity is not near 100%. The other factors, such as low evaporation fraction, seem to be of secondary importance. Earlier the reviewer raised the question about the orange line seeming to be worse after the shape parameter correction. The main reason the shape parameter correction did not have a positive impact was that the shape parameter assumed by the bulk scheme simulation was close to the most common best-fit shape parameter simulated by the bin scheme. In this case, the shape parameter may not have been the most important reason for the two schemes disagreeing, but only because an appropriate shape parameter had been used. When inappropriate values are used (for example the purple and green lines), it is clear that the inappropriate shape parameter was the main cause of the disagreement.

Remove the talk about radiation and ice in the Appendix

We would rather include it for the reasons stated before, but it has been removed.

Responses to Reviewer #3

Thank you to both reviewers for their comments. We have continued to work to clarify the methods and discussion in the manuscript.

Review of study "The role of the Gamma function shape parameter in determining differences between condensation rates in bin and bulk microphysics schemes" authored by A. Igel, and S. van den Heever.

The rates of condensation and evaporation obtained in simulations with bin and bulk microphysical schemes are compared in simulations of non-precipitating shallow cumulus clouds. It is shown that the difference between the rates is largely because of non-optimum choice of shape parameter in the Gamma distribution used in the bulk-scheme. Corrections in the rates of condensation and evaporation in the bulk -scheme are introduced to get better agreement with those in the bin-scheme.

The topic of the paper is important. The calibration of bulk-parameterization schemes using bin-schemes as benchmark is an important way to improve bulk-schemes and the skill of cloud-resolving models. At the same time I have very serious remarks to the current study. The paper cannon be published in the present way. *I would recommend to discuss the possibility of publication after major revision.*

The comments and remarks are the following.

1. General comment: the paper is written in a very unclear way. It is difficult to follow the conclusions and statements of the authors. The paper contains a lot of complicated discussions, assumptions, and conclusions which are not illustrated either by formulas or by figures. We have worked to clarify the discussion throughout the manuscript. In reading through the reviewer's comments, we see that there were some places where the reviewer did not fully understand our analysis methods. In particular it seems that the reviewer thought that we were implementing the shape parameter correction into the model to run new simulations. This was not the case. This misunderstanding probably led to additional confusion throughout the paper. The analysis methods are described more clearly now, and with a clear understanding of our actual methods, some of the arguments that were confusing before should be clear now.

2. line 73. Is it possible to plot in fig 2 (or in a separate figure) the values of shape parameters that can be derived from the bin scheme used?

Yes. We now show example average shape parameter values from some of the joint bins in BIN400, as well as example values of $\overline{f_{NU,BIN}}$. Here is the new figure 2:



Figure 2. (a, b) Example average condensation and evaporation rates (mg kg⁻¹ s⁻¹), (c, d) example average shape parameters, and (e, f) example average values of $\overline{f_{NU,BIN}}$ in joint bins from BIN400. (a, c, e) show average values of the two quantities for all joint bins from BIN400 with *S* between 1.011-1.012 and (b, d, f) show averages for all joint bins from BIN400 with \overline{D} between 19 and 20µm.

3. line 114. Does the expression (2) mean that supersaturation is assumed constant during one time step? I suppose that it is not a good approach, because drop growth and the changes of S are actually described by the same equation. Namely, when droplet grow they immediately decrease S. It is just the mass conservation law.

Supersaturation is not assumed constant during a time step. The RAMS' representation of condensation is actually relatively sophisticated compared to most bulk schemes in that simultaneously accounts for both vapor and heat diffusion to/from hydrometeors. A complete description of the condensation/evaporation is beyond the scope of this paper. We only want to point out the most basic differences between bulk and bin schemes. Based on this comment and many of those that follow, it seems that in our effort to be complete in writing down the condensation equations from the two schemes, we have created more confusion than is necessary. In the revised manuscript, we only indicate the how the condensation rate is proportional to S and the droplet distribution properties without discussing the details of the implementation except where necessary.

4. Most bulk schemes use saturation adjustment, which likely decrease the accuracy of those bulk schemes as compared to that used in RAMS. To what extent the values of corrected factors (eq. (4)) are suitable for other bulk schemes?

The results of this study should not be applied to bulk schemes that use saturation adjustment. This is now explicitly stated in the conclusions.

5. line 116. 1) Eqs (2) and (3) contain very strange notations: r is not the radius (typical notation), but mass mixing ratio. 2) from the notations it is not seen that rc in (2) is cloud water content (CWC), and in Eq. (3) rc is mass content of droplets belonging to the i-th bin in the bin scheme. The utilization of the same notations to different quantities leads to confusion, and leads to the necessity of long explanations in the text. I would recommend to use bin indexes in case the bin scheme is discussed.

We agree that r is often radius, but it is also usually the symbol used for mixing ratio (e.g. the AMS Glossary entry for mixing ratio). In the revised equations, only $\partial r_c/\partial t$ appears, and the definition of this term is explicitly stated.

6. line 116. Most bulk schemes use saturation adjustment, which likely decrease the accuracy of those bulk schemes as compared to that used in RAMS. To what extent the results about the choice of the shape parameter (or corrections implemented in eq. 4) are suitable for other bulk schemes?

Schemes that use saturation adjustment should not be at all sensitive to the choice of shape parameter, and the results of this study will not be applicable to those schemes. This is now made clear in the conclusions.

7. line 125. Table 1 present notations. The table does not present explanations. I suppose the expressions for condensational/evaporation growth should be presented clearer. Yes, we only intended Table 1 to give definitions of the symbols. We have eliminated Table 1 given that we have substantially simplified Eqs. 2 and 3.

8. line 133. What is time step used in BULK in Eq. (2)? The characteristic time scale of the change of S is drop relaxation time during which |S-1| falls trice. Time step should be smaller than the drop relaxation time. Otherwise utilization of the Eulerian integration scheme can lead to RH<100% in case of condensation. (This is the reason of the utilization of substeps in the bin-scheme).

It is the full model time step (1s). Notice in the previous version of the manuscript that Eq. 2 uses $S^{t+\Delta t}$. This makes the equation implicit. Without going into the details, Eq. 2 is a simplified and incomplete version of the actual equations used in the condensation scheme. The methods

used to solve this implicit equation are such that if RH>100%, it will still be \geq 100% at the end of the time step. See Walko et al. (2000) for full details.

9.line 158. It is not clear how do you use the approach to calculate S in the bulk scheme using the approach used in the bin scheme. Do you mean that you used analytic solution for S? How did you calculate coefficients in the equations supersaturations S and Si , which (i.e. coefficients) include size distributions? If you know supersaturation integral, why do you not use the bin-emulating procedure of recalculation of drop masses in each "bulk" bin? We have made this clearer. We are only referring to the calculation of the saturation ratio at the beginning of the microphysics routines. We wanted to make sure that both schemes would diagnosis the same value of the saturation ratio from water vapor mixing ratio and temperature. This was originally not the case. The bulk scheme originally used an empirical formula, and the bin scheme used a formula based on the Clausius-Clapeyron equation.

10. line 172. The shape parameters may change with height because the shape of DSD changes. Sometimes the shape parameter should be changed together with other parameters of Gamma distribution.

Some bulk microphysics schemes do have methods for diagnosing the cloud droplet shape parameter. Our scheme does not. A constant value in time and space must be used.

11. line 186. There is no v in eq (3) Yes, that is correct.

12. line 193. Correct typo. Thank you, it has been corrected.

13. Line 207 It is not clear how calibration can be performed when the bulk and the binschemes produce different droplet concentrations (because of different reasons including differences in aerosol concentrations). If droplet concentrations are different, it means that the DSD shapes in BULK and BIN should be different just because the DSD shape depends on the droplet concentration. It seems to me that it would be better to choose aerosol concentration in BULK in such a way to get similar droplet concentrations in BULK and BIN.

It is because of these concerns that we are binning all of the output by number concentration (and mean diameter and saturation ratio). This way, we only compare cloudy points in from the bulk simulations with cloudy points from the bin simulations that have very similar number concentrations.

14. line 219. Supersaturation of 1% is quite large value. It is not clear why grid points with such and lower values were excluded from the analysis.

These points are no longer excluded from the initial analysis. We also have included a new section where we more clearly show how our results depend on relative humidity and how the initial analysis changes if we exclude points with RH of 99.5-100.5% (rather than 99-101% as we did in the previous version of the manuscript).

15. line 227. Fig. 2 is not clear. What is plotted in the figure? How were these figures obtained? Among many questions concerning this figure: why the condensation or evaporation rates are positive at any RH. Are these diagrams obtained by averaging over cloud volume? Over cloud life time?

Thank you for catching this mistake. This was the wrong plot. It was showing shape parameter, not condensation rate. The correct figure is now included in the manuscript.

16. line 238 In fig 3 "original", but not ORIG.

Thank you. In the revised manuscript, this sentence has been removed.

17. line 317. What are the values of the ratio f_nu , bin/f_nu_bulk? Are these bulk are time and spatial averaged?

At your suggestion, we have included in Figure 2 examples of $\overline{f_{NU,BIN}}$ from BIN400 (see comment #2). These are average values of all cloudy points that fall in each joint bin, regardless of where they occurred in space or time. The values of f_nu,bin/f_nu,bulk will of course depend on which bulk simulation is being compared to each bin simulation. Specifically, $f_{NU,BULK} = 0.69, 0.81$, and 0.88 for NU2, NU4, and NU7, respectively. These values are now specified in the manuscript.

18. If f_nu , bin/f_nu_bulk are calculated for each phase space bin, do you calculate a lookup tables to use in bulk simulations? How would these values depend on the stage of cloud evolution and on cloud parameters (cloud top height). How would these values depend on aerosol concentration?

Can you present tables of these values? The application of formula (4) should be described clearer with examples of size distributions, the fields of CWC, fields of concentration, mean volume radius, etc.

We did not explain clearly what we are doing. We are not rerunning any bulk simulations with information about f_nu,bin/f_nu,bulk, so there is no need to create look-up tables. We are only taking the outputted condensation/evaporation rates from the bulk simulations, adjusting their values using f_nu,bin/f_nu,bulk, and then recomparing to the bin condensation/evaporation rates. Another way to say this is that we are taking our original ln(ratios) (which is different for every joint bin and simulation pair) and multiplying them by f_nu,bin/f_nu,bulk (which is also different for every joint bin and every simulation pair) and looking at how the histograms of these adjusted ln(ratios) change.

19. line 338. Please provide DSD in bulk and DSD in bin before and after correction.

The DSDs themselves do not change. Hopefully this makes sense given the better explanation of our methods.

20. Please present comparison of fields of CWC (and concentrations) in bin and in bulk scheme before and after corrections. Only such comparison can say whether the correction introduced in (4) led to improvement of the bulk scheme.

CWC and droplet concentrations do not change as a result of the correction since we are not running new simulations.

21. line 433. I suppose that it is necessary to compare DSD in bin and bulk schemes. Otherwise it is impossible to understand what were the changes in the DSD in the bulk scheme as a result of correction expressed by eq. (4).

This comment seems to be related to our inadequate description of our methods. There were no changes to the DSD as a result of (4). Hopefully this is clear now.

22. lines 440-444. The discussion is not clear. The changes in the shape (and amplitude) of DSD can be recalculated into the changes condensation/evaporation rates. So, these changes are closely related. Again, what were the changes in DSD predicted by bulk-scheme after correction expressed by (4)?

This section has been substantially revised. As mentioned above, the bulk scheme simulations have not been rerun, so there are no changes to the DSD to discuss.

23. lines 462-472. The conclusions should be formulated better. First, which results of the authors justify that the Gamma distribution is a good assumption of the DSD? I did not find such justifications in the paper.

This conclusion has been removed. We made this statement based on the fact that the NRMSE values were reasonably low, and that by assuming that the BIN simulated gamma DSD's we could get good agreement in terms of condensation/evaporation rates with the BULK simulations.

Second, immediately, the authors state that the exact knowledge of the shape is not necessary. We meant that having the detailed binned distribution information may not be necessary – an assumption of a gamma PDF may be sufficient if the proper shape parameter is known.

Third, immediately after these conclusions, the authors conclude that the shape parameter is responsible for agreement/disagreement with the bin -scheme results. All these statements seem contradict each other. The text should be shortened and rewritten clearer. Thank you. The conclusions have been modified to make these points clearer.

24. line 474. Despite the statement that the shape parameter is the main factor that allows to perform calibration, the procedure expressed by (4) does not correct the shape parameter, but just adjusts condensation/evaporation rates. What is the advantage of such approach vs the correction of the shape parameter itself.

The reviewer is correct in that (4) only adjusts the condensation/evaporation rates based on our knowledge of how the best-fit shape parameters in the bin simulations differ from the assumed shape parameters in the bulk simulations. This is a procedure that we have used in order to demonstrate that the shape parameter is important for the different condensation and evaporation rates predicted by the two schemes. In an actual simulation, we would not want to use such a correction, but would instead want to have the correct value of the shape parameter at every cloudy grid point.

It seems that this factor should depend on aerosol concentration

The distribution of best-fit shape parameters (and therefore also correction factors) that arise in the bin simulations does depend on aerosol concentration. Below is a figure from Igel and van den Heever 2017a showing the distributions of best-fit shape parameters from these simulations.



FIG. 5. Frequency distributions of the best-fit shape parameters. Frequency distributions from BIN100, BIN400, and BIN1600 are shown in blue, red, and yellow, respectively. The different line styles show the distribution using all data (solid), data from supersaturated regions (dashed), and data from subsaturated regions (dotted–dashed).

25. Line 485. The conclusions should be formulated clearer. What the authors propose to do with their bulk-scheme: to multiply the condensation/evaporation rates by some factor? Will this factor tabulated according to certain conditions, cloud stage evolution, etc.? We are suggesting that more work needs to be done to appropriately diagnose or predict the shape parameter in bulk microphysics schemes in order to improve their ability to simulate clouds. The best way to diagnose or predict the shape parameter is not addressed by the paper. We have made this clearer in the revised manuscript.

Relevant Manuscript Changes:

1. Changes throughout the manuscript to clarify the methods, discussion, and conclusions.

2. Tables 1 and 2 have been removed. Table 1 was removed after Eqs. 2 and 3 were simplified and used far fewer symbols, and Table 2 was removed at the suggestion of Reviewer #2.

3. Figure 2 has been corrected, and additional panels have been added to show shape parameter and f_{NU} .

4. Figure 3 has been modified to include the standard deviation data in the legends (instead of in Table 2) and no longer excludes data based on relative humidity.

5. Figures 4 and 5 are new. Figure 4 addresses the dependence of the agreement on relative humidity. Figure 5 repeats the analysis in Figure 3 (Figure 5 is similar to the original Figure 3). New discussion accompanies both figures.

6. Figure 7 replaces the previous Figure 5 and more clearly shows the dependence of the agreement on NRMSE and fractional mass change. Most of the discussion related to these figures has been rewritten.

1	The Role of the Gamma Function Shape Parameter in Determining
2	Differences between Condensation Rates in Bin and Bulk Microphysics
3	Schemes
4	
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17	Abstract. The condensation and evaporation rates predicted by bin and bulk microphysics
18	schemes in the same model framework are compared in a novel-statistical way using simulations
19	of non-precipitating shallow cumulus clouds. Despite other fundamental disparities between the
20	bin and bulk condensation parameterizations, the differences in condensation rates are
21	predominantly explained by accounting for the width of the cloud droplet size distributions
22	simulated by the bin scheme. While <u>T</u> the bin scheme does not always predict a cloud droplet size
23	distribution that is well represented by a gamma distribution function (which is assumed by bulk
24	schemes); however, this fact does not appear to be important appears to be of secondary
25	importance for explaining why the two scheme types predict different condensation and
26	evaporation rates. The width of the cloud droplet size is not well constrained by observations and
27	thus it is difficult to know how to appropriately specify it in bulk microphysics schemes.
28	However, this study shows that enhancing our observations of this width and its behavior in
29	clouds is important for accurately predicting condensation and evaporation rates.

30 **1. Introduction**

31

32	Bin and bulk microphysics schemes are both popular approaches for parameterizing subgrid-
33	scale cloud processes as evidenced by the large number of schemes that have been developed.
34	Tables 2 and 3 in Khain et al. (2015) summarize the characteristics of dozens of microphysics
35	schemes, and discuss in detail the basic principles of the two basic types of schemes. Briefly, in
36	double-moment bulk schemes, the mass mixing ratio and total number mixing ratio for
37	predefined hydrometeor species are predicted, and a function is assumed to describe the shape of
38	the size distribution of each species. In contrast, bin schemes do not assume a size distribution
39	function, but instead, the distribution is broken into discrete size bins, and the mass mixing ratio
40	is predicted for each bin. Usually the size of each bin is fixed, in which case the number
41	concentration is also known for each bin.
42	
43	Bin schemes, particularly those for the liquid-phase, are generally thought to describe cloud
44	processes more realistically and accurately than bulk schemes, and thus they are often used as the
45	benchmark simulation when comparing simulations with different microphysics schemes (e.g.
46	Beheng, 1994; Seifert and Beheng, 2001; Morrison and Grabowski, 2007; Milbrandt and Yau,
47	2005; Milbrandt and McTaggart-Cowan, 2010; Kumjian et al., 2012). For the ice phase, bin

48 schemes are subject to many of the same issues as bulk schemes, such as the use of predefined

49 ice habits (which may not always appropriately describe real-world ice) and the conversion

50 between ice types (the real atmosphere does not have strict categories for ice), rendering them

51 not necessarily more accurate (Khain et al. 2015). Regardless, bBin schemes are much more

52 computationally expensive since many additional variables need to be predicted. As a result, bin

53 schemes are used less frequently. It is of interest then to see how well bulk and the more accurate 54 liquid-phase bin microphysics schemes compare in terms of predicted process rates, and to assess 55 how much predictive value is added by using a bin instead of a bulk microphysics scheme.

56 Furthermore, comparison of process rates in bin and bulk schemes could help to identify ways in

57 which to improve bulk schemes.

58

One of the primary drawbacks of double-moment bulk schemes that assume probability distribution functions (PDFs) is that many microphysical processes are dependent on the distribution parameters that must be either fixed or diagnosed. In the case of a gamma PDF which is typically used in bulk schemes, this parameter is the shape parameter. The gamma size distribution (*n*) is expressed as

64

$$n(D) = \frac{N_t}{D_n^{\nu} \Gamma(\nu)} D^{\nu - 1} e^{-D/D_n}$$
(1)

65 where *v* is the shape parameter, $N_{\underline{t}}$ -is the <u>total</u> number mixing ratio, *D* is the diameter, and 66 D_n is called the characteristic diameter. All symbols are defined in Table 1 for reference. 67 Much is still to be learned regarding what the most appropriate value of th<u>e shape</u> parameter is 68 and how it might depend on cloud microphysical properties.

69

70 Figure 1 shows previously proposed relationships between the cloud droplet number

concentration and the shape parameter (Grabowski, 1998; Rotstayn and Liu, 2003; Morrison and

72 Grabowski, 2007; hereinafter G98, RL03, and MG07, respectively) along with values of the

- shape parameter reported in the literature and summarized by Miles et al. (2000) for several
- 74 different cloud types. The figure shows a wide range of possible values of the shape parameter
- based on observations. The lowest reported value is 0.7 and the highest is 44.6, though this

highest point is clearly an outlier. Furthermore, there is no apparent relationship with the cloud droplet concentration in the data set as a whole, and both increases and decreases of the shape parameter are found with increasing droplet concentration among individual groupings. There is also no clear dependence of the shape parameter on cloud type. Figure 1 additionally shows that two of the proposed functions relating these two quantities are similar (RL03 and MG07), but that the third function (G98) exhibits an opposite trend compared with these first two.

82

83 Furthermore, using appropriate values of the shape parameter may be necessary to accurately 84 model cloud characteristics and responses to increased aerosol concentrations. Morrison and 85 Grabowski (2007) found that switching from the MG07 to the G98 N-v relationships in Figure 1 86 led to a 25% increase in cloud water path in polluted stratocumulus clouds. This example shows 87 that inappropriately specifying the shape parameter could have implications for the accurate 88 simulation of not only basic cloud and radiation properties but also for the proper understanding 89 of cloud-aerosol interactions. However, it is apparent from Figure 1 that *large uncertainties still* 90 exist regarding the behavior of the shape parameter and how it should be represented in models. 91 The goal of this study is to compare the condensation and evaporation rates predicted by bin and 92 bulk microphysics schemes in cloud-resolving simulations run using the same dynamical and 93 modeling framework and to assess what the biggest sources of discrepancies are. The focus is on 94 condensation and evaporation since these processes occur in all clouds and are fundamental for 95 all hydrometeor species. It will be shown that in spite of other basic differences between the 96 particular bulk and bin microphysics schemes examined here, the lack of a prognosed shape 97 parameter for the cloud droplet size distribution in the bulk scheme is often the primary source of

98 differences between the two schemes. Thus, an improved understanding of the shape parameter99 is necessary from observations and models.

100

101 2. Condensation/Evaporation Rate Formulations

102 The Regional Atmospheric Modeling System (RAMS) is used in this study. It contains a double-

103 moment bulk microphysics scheme (BULK) (Saleeby et al., 2004) and the Hebrew University

104 spectral bin model (BIN) (Khain et al., 2004). The Hebrew University spectral bin model is

newly implemented in RAMS. Details about the implementation can be found in Appendix A.

106

107 In the BULK microphysics scheme, condensation/evaporation is treated with a bulk approach. 108 Celoud droplet size distributions are assumed to conform to a gamma probability distribution 109 function (PDF)PDF given by Eq. (1). The condensation/evaporation scheme is described in detail 110 in Walko et al. (2000), and the amount of liquid water condensed in a time step is given by their 111 Eq. 6. Here, a slightly rearranged and simplified version of this equation is presented in order to 112 highlight the similarities to the BIN condensation/evaporation equation shown below. only the 113 important relationships to the cloud droplet distribution properties are shown. Specifically, the 114 BULK condensation/evaporation rate $(\partial r_c/\partial t;$ time rate of change of the mass mixing ratio of 115 cloud droplets) is proportional to N_t , \overline{D} (mass mean diameter), v, and S in the following 116 way:equation is written as $\frac{\partial r_c}{\partial t} \propto (S-1)r_c^{t+\Delta t} = r_c^* + 2\pi \left[N\bar{D}\nu \left(\frac{\Gamma(\nu)}{\Gamma(\nu+3)}\right)^{1/3} f_{\nu,BULK}\right] N_t \bar{D}\nu \left(\frac{\Gamma(\nu)}{\Gamma(\nu+3)}\right)^{1/3} G_{BULK}(S^{t+\Delta t} - S^{t+\Delta t})$ 117 118 1)∆t. (2)119 The BULK scheme does not use a saturation adjustment scheme for cloud water like many other

bulk microphysics schemes do. Also, while not obvious here, the BULK scheme

121 condensation/evaporation is implemented in such a way that evaporation cannot result in 122 supersaturation, and likewise condensation cannot deplete the water vapor so much that the air is 123 subsaturated at the end of the time step. 124 125 In contrast, the equation for the condensation/evaporation rate in the BIN is given by proportional 126 to S, and the number concentration N and diameter D in each bin in the following way: $\frac{\partial r_c}{\partial t} r_{\epsilon}^{t+\Delta t} \propto (S-1) \sum N_i D_i = r_{\epsilon}^* + 2\pi \left(\sum N_i D_i f_{\overline{\nu}i,BIN} \right) G_{BIN} \int_{\Omega}^{\Delta t} (S-1) dt.$ 127 (3) 128 As we would expect in a bin scheme, the condensation rate is proportional to the droplet properties in each bin rather than on the average droplet diameter and total number 129 130 concentration. In the bin scheme, many small sub-time steps are taken during 131 condensation/evaporation and the values of S, N_i , and D_i are updated after each. Semi-analytical 132 equations are used to solve for the time integral of supersaturation that appears at the end of Eq. 133 3 (Khain and Sednev, 1996). In both equations, r_e is the cloud mass mixing ratio, f_{\pm} is the 134 ventilation coefficient, G is a term that accounts for latent heating, vapor diffusion and heat 135 diffusion, S is the saturation ratio, and t is time. The saturation ratio is defined as the ratio of the 136 water vapor partial pressure to the saturated water vapor partial pressure. More details are given 137 in Table 1. 138 139 Although both equations have the same basic form, there are two primary differences in how 140 these equations are formulated: • In the BIN, as is required by the model structure, the condensation rate is calculated for 141

In the BIN, as is required by the model structure, the condensation rate is calculated for
 each bin of the distribution, and these rates are then summed over all bins, as opposed to
 the integration of the gamma distribution that is done in the BULK scheme.

145

• The time step integration is performed semi-analytically in the BIN with multiple subtime steps rather than implicitly in the BULK scheme.

146 These differences between the bin and bulk schemes will be taken into consideration in this

- 147 analysis in order to understand why the two schemes produce different condensation rates.
- 148

149 **3. Simulations**

150 In order to investigate the difference in condensation rates predicted by the two microphysics 151 schemes, simulations of *non-precipitating* shallow cumulus clouds over land were performed. 152 This cloud type was chosen in order to minimize the indirect impacts of precipitation processes. 153 Furthermore, the daytime heating and evolution of the boundary layer results in a wider range of 154 thermodynamic conditions than would occur in simulations of maritime clouds. The wider range 155 of thermodynamic conditions make the conclusions of this study more robust. The simulations 156 were the same as those described in Igel et al.and van den Heever 2016a2017a-b. They were run 157 with RAMS and employed 50m horizontal spacing and 25m vertical spacing over a grid that is 158 12.8 x 12.8 x 3.5 km in size. Such fine spacing was used in order to well resolve the cumulus 159 clouds and their microphysical structure. The simulations were run for 9.5 hours using a 1s time 160 step. Clouds appeared after about 4.5 hours. The simplified profiles of potential temperature, 161 horizontal wind speed, and water vapor mixing ratio based on an Atmospheric Radiation 162 Measurement (ARM) Southern Great Plains (SGP) sounding from 6 July 1997 at 1130 UTC (630 163 LST) presented in Zhu and Albrecht (2003) (see their Fig. 3) were used to initialize the model 164 homogeneously in the horizontal direction. Random temperature and moisture perturbations 165 were applied to the lowest model level at the initial time.

167 Some modifications were made to the model for this study only in order to make the two 168 microphysics schemes more directly comparable. The calculation diagnosis of saturation ratio 169 from current values of the water vapor mixing ratio and temperature at the beginning of the 170 microphysics routines was changed in the BULK scheme to make it the same as the calculation 171 in the BIN. The BIN does not include a parameterization for aerosol dry deposition, so this 172 process was turned off in the BULK scheme. Finally, the regeneration of aerosol upon droplet 173 evaporation was deactivated in both microphysics schemes. Aerosol concentrations were 174 initialized homogeneously in the horizontal and vertical directions. Aerosol particles did not 175 interact with radiation. 176 177 Five simulations were run with the BULK scheme and three with the BIN scheme. Since the 178 relationships in Figure 1 (G98; RL03; MG07) suggest that the shape parameter may depend on 179 the cloud droplet number concentration, the simulations were run with three different aerosol concentrations, specifically, 100, 400, and 1600 cm⁻³, in order to obtain a larger range of droplet 180 181 concentration values. The aerosol in the BIN simulations was initialized with, and in the BULK 182 simulations was assumed to follow, a lognormal distribution with a median radius of 40nm and a 183 spectral width of 1.8. These BULK simulations used a shape parameter value of 4. Two 184 additional BULK simulations were run with an aerosol concentration of 400 cm⁻³ and shape 185 parameter values of 2 and 7. These values were chosen based on previous analysis of the BIN 186 simulations in Igel et al. and van den Heever 2016a2017a. The BIN simulations will be referred 187 to by the microphysics scheme abbreviation and the initial aerosol concentration, e.g. BIN100, 188 and the BULK simulation names will additionally include the value of the cloud droplet shape 189 parameter, e.g. BULK100-NU4.

191 **4. Results**

192 4.1 Instantaneous Condensation Rates

193 In order to compare directly the condensation rates predicted by the BULK and BIN 194 microphysics schemes, it is necessary to evaluate these rates given the same thermodynamic and 195 cloud microphysical conditions. The BULK condensation equation (Eq. (2)) is approximately 196 linearly proportional to four quantities: S, N_t , \overline{D} , and v. We say approximately proportional since 197 the presence of the ventilation coefficient (which itself depends on \overline{D} and v) makes these factors 198 not truly proportional to the condensation rate. In the BIN scheme, among these four variables, 199 the condensation rate is only explicitly proportional to S, and is not explicitly proportional to N_t , 200 \overline{D} , or v (which do not appear at all in Eq. (3)) since the BIN scheme does not make assumptions 201 about the functional form of the size distribution. If it is assumed nevertheless that the BIN size 202 distributions can be described by some probability distribution function (which does not 203 necessarily have to be a gamma distribution), then we would still expect the BIN scheme 204 condensation rate to scale linearly with N_t and \overline{D} .

205

Therefore, in order to best compare the condensation rates between the two schemes, the condensation and evaporation rates that occur during one time step were binned by the values of S, N_t , and \overline{D} that existed at the start of the condensation/evaporation process and were averaged in each bin. (Note that these phase space bins are not the same as the hydrometeor distribution bins.) That is, all points with the same $S, N, \text{ and } S, N_t$, and \overline{D} were grouped and the average condensation or evaporation in each group of points was calculated. The average condensation rate in each S, N_t , and \overline{D} joint bin was calculated separately for alleach simulations.

214	Examples of the average condensation and evaporation rates from BIN400 are shown in Figure
215	2a-b as functions of S, N_t , and \overline{D} . Values in each joint bin differ for the other simulations. Where
216	the cloud was supersaturated or subsaturated, sSaturation ratio bin widths of 0.1 or 1 were used
217	where the cloud was supersaturated or subsaturated,, respectively. For \overline{D} , bin widths of 1 μ m
218	were used. For N , the bin width depended on the initial aerosol concentration of the simulation:
219	bin widths of 2.5, 10, and 40 mg^{-1} were used for simulations with an initial aerosol concentration
220	of 100, 400, and 1600 mg ⁻¹ , respectively. The output from the <u>dynamical</u> model only includes
221	the values of S, $N_{\underline{t}}$, and \overline{D} after condensation and evaporation have occurred. However, since the
 222	rates of condensation and droplet nucleation were known from additional model output, and
223	since microphysics was the last physical process to occur during a time step in RAMS, the S, $N_{\underline{t}}$
224	and \overline{D} that existed before condensation occurred were easily calculated from the model outpetut.
225	
226	Note that the aerosol activation parameterizations in the BULK and BIN microphysics were not
227	the same, and hence the number of nucleated cloud droplets was not the same. This impacted the
228	number of data points within each joint S, N, and \overline{D} bin. However, we are primarily concerned
229	with the average condensation rate in each joint bin, and the average value should not be
230	impacted by the number of data points within a bin provided that the number is sufficiently high
231	(joint bins with fewer than 50 data points are neglected). Therefore, the differences in the aerosol
232	activation parameterizations, or for that matter, differences in the evolution of the cloud fields,
233	should not influence the differences in the average condensation rates as evaluated in our
234	framework.

236	The average condensation rate in each S, N, and \overline{D} joint bin was calculated for all simulations.
237	All points where the cloud mixing ratio before condensation was greater than 0.01 g kg ⁻¹ and the
238	cloud droplet number concentration was greater than 5 mg ⁻¹ were included in the analysis. In
239	addition, grid points with relative humidity between 99% and 101% after condensation or
240	evaporation were excluded. The condensation or evaporation rates at these points was limited by
241	the supersaturation or subsaturation, respectively, and thus the rates were not highly dependent
242	on the droplet characteristics. Finally, joint bins with fewer than 50 data points were discarded.
243	
244	Figure 2 shows an example of the average condensation and evaporation rates in the joint bins
245	for one simulation. As is seen in Figure 2 <u>a-b</u> , there is a smooth transition to higher condensation
246	rates as the saturation ratio increases, and to higher condensation (S \geq 1) and evaporation (S $<$ 1)
247	rates as the diameter or number mixing ratio increases. This is expected based on the
248	condensation equations (Eqs. (2), (3)). All other simulations behave similarly.
249	
250	Note that the aerosol activation parameterizations in the BULK and BIN microphysics were not
251	the same, and hence the number of nucleated cloud droplets was not the same. This impacted the
252	number of data points within each joint S, N_t , and \overline{D} bin. However, we are primarily concerned
253	with the average condensation rate in each joint bin, and the average value should not be
254	impacted by the number of data points within a bin provided that the number is sufficiently high
255	(joint bins with fewer than 50 data points are neglected). Therefore, the differences in the aerosol
256	activation parameterizations, or for that matter, differences in the evolution of the cloud fields,
257	should not influence the average condensation rates as evaluated in our framework.
 258	

259	In order to compare easily the condensation rates predicted by the two microphysics schemes, we
260	calculate the ratio of the average condensation/evaporation rate of each joint bin from a BULK
261	simulation to the average condensation/evaporation rate of the corresponding joint bin from a
262	BIN simulation, and then calculate the natural logarithm of each ratio. These will be referred to
263	as 'ln(ratios)'. logarithm of the BULK to BIN condensation and evaporation rate ratios (these
264	values will be referred to as 'ln(ratios)')We find the ln(ratios) of average
265	condensation/evaporation rate for five pairs of simulations. Specifically, BULK400-NU2,
 266	BULK400-NU4, and BULK400-NU7 are all compared to BIN400, while BULK100-NU2 is
267	compared to BIN100 and BULK1600-NU2 is compared to BIN1600. Histograms of the
268	ln(ratios) this ratio for all pairs of simulations are shown in Figure 3a-b and Figure 3e-f. This set
269	of In(ratio) histograms will be referred to as ORIG. The data have been separated into
 270	subsaturated (evaporating) and supersaturated (condensing) points. Positive values indicate that
271	the rates in the BULK scheme are larger, and negative values indicate that the rates in the BIN
272	scheme are larger. Values of $\pm 0.1 \ (\pm 0.2)$ correspond to about a 10% (20%) difference in the
273	condensation or evaporation rate between the two schemes for the joint bin.
 274	
275	First we examine the impacts of increasing aerosol concentrations on the agreement of
276	evaporation and condensation rates for in BULK and BIN simulations with the same shape
277	parameter. Figures 3a-b show the histograms of the condensation and evaporation rate ln(ratios)
278	for BULK100-NU4 compared to BIN100, BULK400-NU4 compared to BIN400, and
279	BULK1600-NU4 cmopared to BIN1600pairs of simulations with a cloud droplet shape
280	parameter of 4 but with differing initial aerosol concentration. Figure 3a-3b reveals that in
 281	general the condensation rate is higher in the BIN scheme simulations as indicated by the more

frequent negative ln(ratios)., On the other hand, whereas the evaporation rates are more similar between the two schemes as indicated by the most frequent ln(ratios) being equaaul to or slightly greater than 0 in Figure 3a. The evaporation rates are more frequently greater in the BULK scheme simulations. For the simulation pair with an initial aerosol concentration of 1600 cm⁻³, there is a long tail of positive ln(ratio) values. As a result, this pair of simulations has the highest standard deviation of the ln(ratio) values of all simulation pairs (Table 2a).

289 Figures 3e-f show the histograms of condensation and evaporation rate ln(ratios) for the three 290 BULK400 simulations that have with different values of the cloud droplet shape parameter, all 291 compared to BIN400. All three BULK400 simulations are compared to the BIN400 simulation. 292 For both condensation and evaporation, the peak of the ln(ratios) histograms increase as the 293 cloud droplet shape parameter used in the BULK400 simulations increases. For the BULK400-294 NU2 simulation, the condensation and evaporation rates are frequently 20% lower than the 295 BIN400 rates or more whereas for the BULK400-NU7 simulation, the condensation rates 296 compared to the BIN400 simulation are most frequently very similar (ln(ratios) near zero). Thus 297 the value of the cloud droplet shape parameter chosen for use in a simulation is clearly important 298 for determining how well a bulk microphysics scheme compares to a bin microphysics scheme in 299 terms of predicted condensation and evaporation rates.

300

4.2 Accounting for the Shape Parameter

302 Fortunately, we know theoretically how the cloud droplet shape parameter will alter

303 condensation and evaporation rates and this dependency can be accounted for in our comparison

304 of the two microphysics schemes. The shape parameter term in Eq. (2) (hereafter f_{NU}), which is

equal to $\nu \left(\frac{\Gamma(\nu)}{\Gamma(\nu+3)}\right)^{1/3}$, indicates that when a gamma PDF is assumed, the condensation rate is 305 306 proportional to the shape parameter v such that a higher shape parameter results in higher 307 condensation rates. Of course, T the BIN scheme makes no assumptions about the size 308 distribution functionality and its condensation scheme does not depend on the shape parameter. 309 However, in order to characterize the shape of the predicted BIN cloud droplet size distributions, 310 and to facilitate the comparison of the BIN and BULK condensation rates, we assumed that the 311 predicted BIN size distributions are gamma PDF-like and found the best-fit gamma PDF 312 parameters (see Eq. (1)) for the cloud droplet size distributions at every cloudy grid point in the 313 BIN simulations. We then evaluated the mean value of f_{NU} using these best-fit shape parameters 314 for each joint bin in the S, N, and \overline{D} phase space. 315 316 In order to find the best-fit shape parameters, we defined cloud droplets as belonging to one of 317 the first 15 bins of the BIN liquid array (the remaining 18 bins contain raindrops), which 318 corresponded to a maximum cloud droplet diameter of 50.8 µm. Many methods are available to 319 find such best-fit parameters, but they generally all give similar results (McFarquhar et al., 320 2014). Here we used the maximum-likelihood estimation (MLE) method. For our problem, the 321 log-likelihood function $(\ln(L))$ is defined as $ln L = \frac{1}{N_t} \sum_{i=1}^{15} N_i \ln n(D_i) \prod_{i=1}^{15} N(bin_i) \ln n(bin_i)$ (4) 322 where N(bini) is the simulated number concentration of cloud droplets in the ith bin of the liquid 323 324 size distribution array (each bin corresponds to a particular droplet diameter) and $n(bin_i)n(D_i)$ is 325 the value of the gamma PDF as defined in(-Eq. 1) for D_i with unknown values of the parameters 326 D_n and v. The function is normalized by the total cloud droplet concentration N_t in order to \$27 remove N_t as a free parameter in Eq. 1. As indicated by its name, the MLE method seeks to

maximize the log-likelihood function given by Eq. 4. To do so, we used the MATLAB function fmincon to find the parameter values that minimized -1*L and found best fits that minimize the error in the total number concentration. Using this method, the size distributions were first normalized by the corresponding total number concentration, leaving only D_{μ} and ν as free parameters of the distribution (Eq. 1).

333

334 Note that while we could determine the values of S, N_t , and \overline{D} that existed before condensation 335 occurred, we could not determine the value of the best-fit shape parameter for this time because 336 the change in mixing ratio of each bin was not output by RAMS. Thus, the average shape 337 parameters used in the analysis are those that exist at the end of the time step. Nonetheless, given 338 the short time step used in these simulations, it was not expected that the best-fit shape parameter 339 would change much in one time step in most cases. The exception may be for very broad 340 distributions characterized by low shape parameters. In part due to this concern, cloudy points 341 with best-fit shape parameters less than 1 are not included in the analysis. This criterion 342 eliminated 4.5%, 5.1%, and 8.6% of the data in BIN100, BIN400, and BIN1600, respectively. 343 Overall, the impact of using the post-condensation shape parameters is not expected to have a 344 large impact on the results. Examples of the average shape parameters in each joint bin are 345 shown in Figure 2c-d. The shape parameter tends to increase with droplet concentration and be 346 low (5 or less) for relative humidity less than 99%. In depth analysis of the best-fit shape 347 parameter in the BIN simulations is found in Igel and van den Heever (2017a). 348 349 Using these best-fit shape parameters from the BIN simulations and the specified shape

parameters from the BULK simulations, \mathbf{T} the shape parameter term (f_{NU}) can be evaluated for

351	each cloudy point for all simulations. joint bin in the S, N, and \overline{D} phase space for all simulations.
352	In the case of each BULK simulations, the value of $f_{NU,BULK}$ for the same for every joint
353	bincloudy point since the value of $f_{NU,BULK} f_{NU}$ is uniquely determined by the choice of the shape
354	parameter value for each BULK simulation. Specifically, $f_{NU,BULK} = 0.69, 0.81$, and 0.88 for
355	<u>NU2, NU4, and NU7 simulations, respectively.</u> For the BIN simulations, $f_{NU,BIN}$ f _{NU} -can be
356	calculated using the best-fit shape parameters and will have a different value for every cloudy
357	grid point. The values of $f_{NU,BIN}$ for the cloudy grid points in each joint bin were averaged
358	together to find a mean $\overline{f_{NU,BIN}}$ for each joint S, N_t , and \overline{D} bin for each BIN simulation. Example
359	values of $\overline{f_{NU,BIN}}$ for some joint bins are shown in Figure 2c-d2e-f. Unlike for the BULK
360	simulations, the value of f_{NU} for the BIN simulations will vary amongst the joint bins since the
361	best-fit shape parameter is determined from the freely evolving cloud droplet distributions that
362	are predicted by the BIN microphysics scheme. We can use the values of $f_{NU,BULK} f_{NU}$ and
363	$\overline{f_{NU,BIN}}_{in our comparison of the condensation and evaporation rates to account for the$
364	differences in condensation and evaporation rates between the two schemes that arise due to
365	different size distribution widthsshape parameters the fact that the best-fit shape parameters in the
366	BIN simulations will often be different from the single prescribed value in the BULK
367	simulations. Specifically, in our analysis, we adjusted the mean condensation and evaporation
 368	rates (<i>C</i>) for each joint bin from the BULK simulations in the following way:
369	$\overline{C_{BULK,corrected}} = \overline{C_{BULK,original}} \frac{\overline{f_{NU,BIN}} f_{NU,BIN}}{f_{NU,BULK}} $ (45)
370	Note again that the value of $\overline{f_{NU,BIN}} f_{NU,BIN}$ will be different for each joint bin. By making this

371 correction, we found the condensation and evaporation rates that the BULK simulations *would*

have had if they had used the same value of the shape parameter that best characterized the cloud

droplet size distributions that were predicted by the BIN simulations. <u>To be clear, we did not run</u>
<u>new simulations, rather the outputted condensation/evaporation rates from the existing BULK</u>
simulations were adjusted for the purposes of our analysis using Eq. 45 to account for the
<u>differences in size distribution shapes between the BIN and BULK simulations. We will next</u>
<u>compare these adjusted BULK condensation/evaporation rates to the BIN rates to see if the</u>
<u>comparison improves.</u>

379

380 The ln(ratios) of the modified adjusted condensation and evaporation rates from the BULK 381 simulations to the rates from the BIN simulations are shown in Figures 3c-d and Figures 3g-h. 382 Hereafter, these ln(ratios) will be called adjusted ln(ratios). This set of ln(ratios) will be referred 383 to as CORR. The most frequent value of the CORR adjusted - ln(ratios) is near zero (indicating 384 that the two schemes predict the same rate) for all simulation pairs and for both condensation and 385 evaporation. The impact of the modification adjustment is most notable in Figures 3g-h where 386 the histograms of the CORR adjusted ln(ratios) now nearly lie on top of one another whereas in 387 Figures 3e-f they are clearly separated. Thus, it appears that our method of accounting for the 388 value of the shape parameter has worked well.

389

Furthermore<u>Additionally</u>, the standard deviations of the condensation rate CORR adjusted
ln(ratio) histograms (shown in the legend of each panel) for condensation is are decreased by
about half compared to the ORIG ln(ratio) histograms (Table 2a b)slightly. This is not the case
for the evaporation rate CORR adjusted ln(ratio) histograms for evaporation, where forin four out
of five all simulation pairs the standard deviation is increased compared to the ORIG original
ln(ratio) histograms. Nonetheless, given that all CORR adjusted histograms (Fig. 3c-d, g-h) now

have a modal value near 0, whereas this was not the case with the ORIG-original histograms
(Fig. 3a-b, e-f), the shape parameter appears to be the primary reason why the condensation and
evaporation rates in the two schemes do not always agree.

- 399
- 400 **4.3 Other Considerations**

While the shape parameter appears to be the primary cause of differences in condensation
and evaporation rates in bin and bulk microphysics schemes, it is worth investigating whetherich
other factors are important.

404

4054.3.1 Relative Humidity

406 When the relative humidity is close to 100%, the condensation and evaporation rates 407 should be are limited by the small supersaturation or subsaturation. In these situations, the droplet 408 properties are expected to have little impact on the condensation or evaporation rate. Instead, 409 these rates will be largely determined by how the schemes behave when the time scale for 410 condensation or evaporation is smaller than the time step of the model. Figure 4 shows the 411 average and standard deviation of the adjusted ln(ratios) for all five pairs of simulations as a 412 function of relative humidity. Both the average and the standard deviation peak for relative 413 humidity near 100%. This indicates that the agreement between the bulk and bin schemes on 414 condensation/evaporation rates scheme is poor, just as we expected it to be based on the above 415 arguments. That said, condensation and evaporation rates occurring with relative humidity near 416 100% are small in magnitude, and disagreements here are not expected to have a large impact on 417 the simulation evolution. treat

418	We repeated the analysis shown in Figure 3, but excluding data points where the relative
419	humidity before condensation/evaporation was between 99.5% and 100.5%. The results are
420	shown in Figure 5. Qualitatively, the results in Figures 3 and 5 are similar. The adjusted
421	histograms are all centered near 0, but The reduction in the decrease in the standard deviation of
422	the ln(ratios) (shown in the legends) from Figure 3 to Figure 5 is substantial, particularly for
423	condensation. This indicates that by removing cloudy points with relative humidity between
424	99.5% and 100.5%, the agreement between the two schemes increases. That said, the standard
425	deviations of the correction adjusted evaporation histograms are still higher than those of the
426	originalunadjusted histograms. Finally, unlike in Figure 3, After the shape parameter correction
427	is applied (Fig. 5c, d, g, h), the standard deviation for the adjusted condensation histograms is
428	consistently lower than that of the adjusted evaporation histograms. Thus Overall, it seems that
429	the correction based on the shape parameter for condensation is more successful than that for
430	evaporation in terms of the spread of ln(ratios). Potential reasons for this difference are explored
431	<u>next.</u>
432	
433	
434	4.3.1-2 Appropriateness of the Gamma PDF and Fractional Mass Change
435	One potential factor reason worth considering is that the gamma PDF is not always
436	appropriate for characterizing the cloud droplet size distributions in the BIN simulations. The
437	BIN microphysics scheme is capable of predicting any shape for the cloud droplet size
438	distributions, including size distributions that may be bimodal. To assess how well our fitted
439	gamma PDFs approximated the actual simulated cloud droplet size distributions, we calculated
440	the normalized root mean square error (NRMSE) of the fits using MATLAB's goodnessOfFit
1	

441 function. An NRMSE of 1 indicates that the fit was no better than a straightflat line equal to the 442 mean of the datasize distribution, and a value of 0 indicates a perfect fit. Figures 4a6a-b show 443 cumulative histograms of the NRMSE values from the three BIN simulations for both 444 evaporating and condensing cloudy points. Note that these are not cumulative histograms of 445 mean values from joint bins as in Figure 3 but rather they are cumulative histograms of the 446 NRMSE values at all individual cloudy grid points in the BIN simulations. The majority of grid 447 points have NRSMSE values between about 0.4 and of 0.6 or lower which indicates that in 448 general the gamma PDF characterizes the simulated cloud droplet size distributions very 449 moderately well. The cumulative distributions of NRMMRSE are similar for all three BIN 450 simulations and similar for evaporating and condensing cloudy grid points. This suggests that the 451 NRMMRSE probably cannot explain why the correction in Figure 5 leads to a reduction in the 452 standard deviation of ln(ratios) for condensation but an increase in the standard deviation of 453 ln(ratios) for evaporation. Nonetheless, we still expect that higher NRMSE should result in 454 differences between the condensation and evaporation rates in bin and bulk schemes. This will 455 be discussed further below.

456

We repeated the calculations of mean condensation or evaporation rate in each *S*, *N*, and \overline{D} joint bin for the BIN simulations, but now we only included those cloudy points with an NRMSE of 0.6 or more (those points with a poor gamma PDF fit). The joint bins for the BULK simulations were unaltered, but did include the modification described by Eq. (4) which now used values of $f_{NU,BIN}$ based only on the high NRMSE points. The resulting histograms of condensation and evaporation rate ln(ratios) are shown in Figures 5a-b for all simulation pairs. The associated standard deviations are listed in Table 2c. This set of histograms will be referred to as CORR- POOR. For evaporation, the peaks of the CORR-POOR ln(ratios) histograms shift to positive
values (Fig. 5a) indicating that the agreement between the BULK and BIN rates is degraded,
although the standard deviations of these histograms are similar compared to the CORR
histograms (Table 2c compared to Table 2b). The shift in peak ln(ratios) suggests that when the
BIN simulations produce cloud droplet size distributions that poorly conform to a gamma PDF,
the best-fit shape parameter is less useful for understanding the differences between BULK and
BIN evaporation rates.

471

472 However, for condensation rates, the results are less clear. Figure 5b shows that many of the high 473 CORR-POOR In(ratio) histograms are still centered near 0, which indicates that the BIN and 474 modified BULK condensation rates still agree well. Furthermore, the standard deviation of these 475 histograms is similar those of the CORR histograms (Table 2b-c). Unlike for evaporation, these 476 results for condensation suggest that the fact that the BIN simulations do not predict cloud 477 droplet size distributions that are similar to gamma PDFs is not an important reason why the 478 BULK and BIN schemes predict different condensation rates. It is unclear why the comparisons 479 of condensation and evaporation rates behave so differently. This uncertainty will be explored 480 next.

481

482 <u>Another 4.3.2 Fraction of Cloud Mass Evaporated</u>

One potential reason that evaporation comparison is generally worse than the<u>and</u> condensation comparisons are different relates to the fractional change of mass. Specifically, the comparison may be better for situations in which only a small fraction of the total cloud droplet mass is condensed or evaporated or condensed within a time step versus a situation in which a large

487 fraction of mass is evaporated or condensed. The reason the fractional change in mass may be 488 important is related to the different treatments of the time step during condensation/evaporation 489 in the two schemes, is that t The BIN microphysics scheme takes an iterative approach to 490 condensation and evaporation in which many small steps are taken. After each small step the 491 droplet properties are updated. When the droplet properties are changing rapidly, this approach 492 may be important for accurately predicting the evolution of the total mass and number of cloud 493 droplets. On the other hand, the RAMS bulk scheme takes just one step (which is equal to the 494 full model time step length) and cannot account for rapidly changing droplet properties within 495 the time step.

496

497 Cumulative histograms of the fraction of cloud mass evaporated in one full time step is shown in 498 Figure 4c-6c for the BIN simulations. Higher fractions of mass are evaporated more frequently as 499 the initial aerosol concentration increases. This result is not surprising given that the high 500 numbers of cloud droplets nucleated from the high numbers of aerosol particles will induce on 501 average higher evaporation rates (Eq (2) and Eq (3)) that cause a higher fraction of mass to be 502 evaporated in one time step. Similarly, cumulative histograms of the fraction of cloud droplet 503 mass condensed in the time step are shown in Figure 446d. Again, high fractions of cloud mass 504 are condensed more frequently as the initial aerosol concentration increases. OverallIn general, 505 large fractional changes in the cloud mass are more frequent during evaporation during 506 condensation. This suggests that the fractional mass change may be a reason for the better 507 comparison of condensation rates than evaporation rates in Figure 5 after the shape parameter 508 correction was applied. 509

510	To explore simultaneously the impact of NRMSE and fractional mass change on the comparison
511	of bin and bulk scheme condensation and evaporation rates, we also calculated the mean
512	NRMSE and fractional mass change of each of the joint S, N_t , and \overline{D} bins in addition to the
513	adjusted mean-ln(ratio) for each bin that we have shown previously. In this analysis, we have
514	excluded points with relative humidity between 99.5% and 100.5% based on our previous
515	analysis of the impact of relative humidity. Joint bins with similar mean NRMSE and fractional
516	mass change were grouped together to find aand the mean of the adjusted mean-ln(ratios) for
517	each group was calculated. Joint bin pairss from all simulation pairs were included. The results
518	are shown in Figure 7, again for condensation and evaporation separately, where colors show the
519	mean of the adjusted mean-ln(ratios) as a function of NRMSE and fractional mass change.
520	Colors near zero (teal) indicate that the two schemes agree well after the shape parameter
521	correction is applied, whereas colors away from zero (blue and yellow) indicate that the two
522	schemes do not agree well even after the shape parameter correctionadjustment is applied.
523	
524	Evaporation will be considered first (Fig. 7a). For fraction of mass evaporated evaporated mass
525	fraction less than about 0.3, the mean adjusted mean-ln(ratios) are near zero. As the fraction of
526	mass evaporated evaporated mass fraction increases above 0.3, the NRMSE also begins to
527	increase, which makes it difficult to understand the influence of either the NRMSE or evaporated
528	mass fraction on the scheme comparison by looking at them in isolation. However, by looking at
529	them together in Figure 7a, we see that the evaporated mass fraction seems to be driving the
530	increase in the adjusted mean ln(ratio) away from 0, particularly when the evaporated mass
531	fraction is greater than 0.4. For these values, the contour lines are approximately flat, which
532	indicates that there is little dependence of the mean adjusted ln(ratios) on NRMSE.

533	
534	The NRMSE seems to be more important for condensation than evaporation. As the NRMSE
535	increases above about 0.5 in Figure 7b for condensation, the mean adjusted mean-ln(ratios)
536	begins to drop away from zero,- and the two schemes have worse agreement on the condensation
537	rateswhich indicates that the bin scheme is predicting higher condensation rates than the bulk
538	scheme. Like for evaporation, when NRMSE and the condensed mass fraction are both relatively
539	low, the mean adjusted ln(ratios) are near zero and show little dependence on NRMSE or
540	fractional mass change.
541	Again, the calculations of mean evaporation rate in each S, N, and \overline{D} joint bin for both the BULK
542	and BIN simulations were repeated but this time with cloudy points separated by low and high
543	mass fraction change. High evaporated mass fraction is defined as 0.25 or higher. Very few
544	cloudy points undergoing condensation have a mass fraction change of 0.25 or higher. Likewise,
545	very few evaporating cloudy points in BIN100 exceed this threshold. Thus, the analysis is only
546	performed for the subsaturated, evaporating cloudy points for simulations pairs that include
547	BIN400 or BIN1600.
 548	
549	The evaporation rate ln(ratio) histograms for the two groups (referred to as CORR-LFR and
550	CORR-HFR) are shown in Figures 5c-d and the associated standard deviations are listed in Table
551	2d-e. It is immediately obvious that the two microphysics schemes behave quite differently for
552	the case of high evaporated fractions. The standard deviation of the CORR-HFR ln(ratio)
553	histograms are up to twice as large as those for ORIG or CORR-LFR (Table 2a,d). Furthermore,
554	most of the CORR-HFR histograms are shifted almost entirely to the right of 0. This result
555	indicates that when the BIN simulations evaporate a high fraction of the cloud mass in one time

step, they almost always predict a higher evaporation rate than the BULK simulations when
given the same initial cloud properties and relative humidity.

558

559 Finally, we found that grid points at which a high fraction of cloud mass is evaporated, the cloud 560 droplet size distributions predicted by the BIN simulations are more likely to fit poorly to a 561 gamma PDF (not shown). Thus, we performed the BULK to BIN evaporation rate comparison 562 twice more: firstly where only BIN simulation points with a high NRMSE of the fitted gamma 563 distributions and a low fraction of cloud mass evaporated were included, and secondly with the 564 opposite conditions where only BIN simulations points with a low NRMSE and a high 565 evaporated fraction were included. The standard deviations of the resultant histograms are listed 566 in Table 2f-g. In the case of high NRMSE and low evaporated fraction, the standard deviations 567 are similar to those for CORR (Table 2b,f), whereas in the case of low NRMSE and high 568 evaporated fraction the standard deviations are high and are similar to those for CORR-HFR. 569 Thus, it seems that the occurrence of high evaporated fraction is more important for explaining 570 poor agreement between the BULK and BIN microphysics scheme than is a poor fit of a gamma 571 PDF to the cloud droplet size distributions simulated by the BIN scheme. 572

573 **5. Conclusions**

574 In this study, we have compared the cloud condensation rates predicted by a bulk and a bin 575 microphysics scheme in simulations of non-precipitating cumulus clouds run using the same 576 dynamical framework, namely RAMS. The simulations were run with three different background 577 aerosol concentrations in order to consider a large range of microphysical conditions. Two

578	additional simulations with the RAMS bulk microphysics scheme were run with different
579	settings for the cloud droplet shape parameter.

581	When the condensation and evaporation rates were binned by saturation ratio, <u>cloud</u> droplet
582	number concentration, and mean diameter, the BULK rates were on average higher or lower
583	depending <u>primarily</u> on the value of the shape parameter used in the BULK simulations. <u>Since</u>
584	the theoretical relationship between the shape parameter and condensation/evaporation rates is
585	known, we adjusted the BULK rates to be those that the simulations would have predicted if they
586	had used the same value of the shape parameter as was found by fitting gamma PDFs to the BIN
587	droplet size distribution output. After doing so, we showed that the BULK and BIN rates were in
588	general in much better agreement, although the condensation rates agreed better than the
589	evaporation rates. After mathematically accounting for the fixed shape parameter assumed for
590	BULK cloud droplet size distributions, we showed that the BULK and BIN rates were in general
591	in much better agreement, although the condensation rates agreed better than the evaporation
592	rates.
593	
594	Other factors were also suggested to impact the agreement of condensation and evaporation rates
595	in the BIN and BULK simulations. First, the agreement was worse as the relative humidity
596	approached 100%. Second, the when the simulated binned size distributions did not conform
597	closely to a gamma PDF (NRMSE was high), the agreement was also worse, particularly for
598	condensation. Lastly, when Additional analysis supported the following conclusions:
1	

A gamma probability distribution appears to be a good assumption for the cloud droplet
 distribution shape, and the exact knowledge of the distribution shape in a bin scheme is often not
 necessary to minimize errors in the condensation rate in bulk schemes.

602 When a large fraction of the cloud droplet population mass wais evaporated or condensed within

a model time step, <u>the agreement was also worse</u>, <u>particularly for evaporation</u>. We hypothesize

604 that the reason for a dependence on the fractional mass change is related to the different

approaches taken by the BIN and BULK schemes to solve the condensation equation. the BIN

606 scheme usually predicts lower evaporation rates than the BULK scheme. This appears to be one

607 reason why the evaporation rates comparison is poorer than the condensation rates comparison. It

608 is possible that the multiple sub-time steps taken by the BIN scheme may be important for

609 accurately predicting evaporation rates. Such a time-stepping approach could easily be

610 implemented in a BULK scheme. This reason for disagreement between the two schemes,

611 however, is of secondary importance compared to the shape parameter. However, all three of

612 these factors were found to be of secondary importance compared to the shape parameter.

613 2.

614 Again, it appears that when the relative humidity is not near 100%, the most important factor for 615 agreement in cloud droplet condensation and evaporation rates between bin and bulk schemes is 616 the shape of the cloud droplet size distribution. Therefore, we feel that MmoreMore effort is 617 needed to understand the behavior of the cloud droplet shape parameter in order to improve the 618 representation of cloud droplet size distributions in bulk microphysics schemes. Improvement in 619 the representation of size distributions should lead to better agreement in the simulated 620 macroscopic properties of clouds by the two schemes, although such potential for better 621 agreement has not been shown here. Finally, while the methods we have used to here to

622	demonstrate the importance of the shape parameter were effective, we are not suggesting that the
623	same methods would be best for improving bulk schemes. and ultimately improve the
624	simulations of clouds.
625	
626	Although we have only investigated two specific schemes, it is expected that the results can be
627	applied more generally to bulk-bin and bulk in schemes that do not use saturation adjustment.
628	Additional work should be conducted using a similar approach in order to compare and evaluate
629	additional microphysics schemes and additional microphysical processes. While it is clear that
630	the effective shape parameter in the bin simulations explains much of the discrepancies in
631	predicted condensation rates between bin and bulk schemes, our understanding of what the most
632	appropriate value of the shape parameter is or how it should vary as a function of basic cloud
633	properties is limited. More work then is also needed toon understanding cloud droplet
634	distribution widths from observations and measurements.
635	

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642

643 Appendix A

644 Implementation of the Hebrew University BIN scheme into RAMS

646	While the present study is only concerned with warm phase processes, the methods to interface
647	the Hebrew University BIN scheme with the RAMS radiation scheme (Harrington, 1997) will be
648	described here, including those for the ice species. The RAMS radiation scheme uses pre-
649	computed lookup tables for the extinction coefficient, single-scattering albedo, and asymmetry
650	parameter for each hydrometeor species. Three of the hydrometeor species in the BIN
651	correspond directly to species in the RAMS microphysics scheme, namely, aggregates, graupel,
652	and hail. All liquid drops are represented as one species in the BIN, so these liquid bins are
653	classified as either cloud droplets or rain drops using the same size threshold used by the RAMS
654	microphysics scheme to distinguish these two species. Finally, the BIN represents three ice
655	crystal types plates, columns, and dendrites. Separate RAMS radiation look-up tables already
656	exist for these different ice crystal types, but like for cloud and rain, there are two tables for each
657	crystal type depending on the mean size of the crystals. In RAMS, the small ice crystals are
658	referred to as pristine ice, and the large ice crystals as snow. Again, the same size threshold used
659	to distinguish these two ice categories is used to assign bins from the BIN ice crystal species as
660	either pristine ice or snow. This fortuitous overlap in the ice species has allowed for the
661	seamless integration of the BIN hydrometeor species with the RAMS radiation scheme. For each
662	set of BIN bins that corresponds to a RAMS species, the total number concentration and mean
663	diameter is calculated, a gamma distribution shape parameter of 2 is assumed, and the
664	appropriate set of look-up tables for the corresponding RAMS species is used for all radiative
665	calculations.

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	(a) Original, all data	(b) Corrected, all	(c) Corrected, high NRMSE	(d) Corrected,	(e) Corrected, high fraction	(f) Corr high NF
	(ORIG)	data (CORR)	only (CORR- POOR)	low fraction mass	mass evaporated	and low fraction
				evaporated (CORR-LFR)	(CORR-HFR)	evapora
Evaporation						
BULK100- NU4/BIN100	0.032	0.025	0.025	-	-	-
BULK400- NU4/BIN400	0.044	0.055	0.056	0.041	0.056	0.038
BULK1600- NU4/BIN160 0	0.097	0.120	0.134	0.090	0.160	0.105
BULK400- NU2/BIN400	0.041	0.05 4	0.053	0.053	0.046	0.041
BULK400- NU7/BIN400	0.061	0.072	0.064	0.047	0.087	0.041
			Conde	nsation		
BULK100- NU4/BIN100	0.057	0.033	0.027	-	-	-
BULK400- NU4/BIN400	0.056	0.027	0.035	-	-	-
BULK1600- NU4/BIN160 0	0.057	0.033	0.032	-	-	-
BULK400- NU2/BIN400	0.059	0.029	0.032	-	-	-
BULK400- NU7/BIN400	0.050	0.026	0.023	-	-	-





2 Figure 1. Shape parameter (*v*) values as a function of cloud droplet concentration as



4 16 previous studies. Values, cloud classification, and groupings are based on their Tables 1 5 and 2. The three solid gray lines show proposed relationships between the cloud droplet 6 concentration and the shape parameter. G98 is from Eq. 9 in Grabowski (1998). RL03 is 7 from Eq. 3 in Rotstayn and Liu (2003) with their α =0.003. MG07 is from Eq. 2 in Morrison 8 and Grabowski (2007). All equations were originally written for relative dispersion, which 9 is equal to $v^{-1/2}$, and have been converted to equations for v for this figure.



17 BIN400 with \overline{D} between 19 and 20 μ m.



Figure 2. The average condensation and evaporation rates (g kg⁻¹ s⁻¹) in joint bins from
BIN400. (a) Joint bins where the relative humidity is 101-101.1% (b) Joint bins where the
cloud droplet diameter is 18-19 μm. (c) Joint bins where the cloud droplet concentration is
20 20-21 mg⁻¹. See the text for more information about the joint bins.



25 Figure 3. The ratio of the BULK to BIN (a-c) condensation and (d-f) evaporation rates as a

24

26 <u>function of saturation ratio (S) and integrated diameter (ND</u> for each pair of simulations.

27 <u>Note the differences in axes limits.</u>



29 Figure 3. Normalized histograms showing the logarithm of the ratio of BULK to BIN (a, c, e,



- 31 original data, and (c-d) and (g-h) show histograms where the correction in Eq. (4) has been
- 32 applied.





38 Figure 5. Like Figure 3, but excluding grid points from the joint bins with relative humidity

39 <u>between 99.5% and 100.5%.</u>







57 evaporated or condensed was calculated. Each panel shows the relationship between the

58 mean NRMSE, mean adjusted ln(ratio) (colors), and (a) mean fraction of mass evaporated

59 or (b) mean fraction of mass condensed. Joint bins from all simulation pairs are included in

High NRMSE: Condensation High NRMSE: Evaporation (a) (b) Norm. Freq. 0.3 0.3 0.2 0.2 0.1 0.1 0 0 -0.4 0.2 -0.4 -0.2 0.2 -0.2 0 n Low Fraction Evaporated **High Fraction Evaporated** 0.4 0.4 0.3 Norm. 0.2 Norm. 0. (c) (d) 0.3 0.2 0.1

0.2

60 <u>the mean adjusted ln(ratios) that are shown.</u>

0

61

-0.4

-0.2

0

In(BULK/BIN Rates)

Figure 5. Similar to Figure 3. Histograms of the logarithm of the ratio of BULK to BIN
condensation and evaporation rates but with conditional sampling of the data. (a-b) Only
BIN simulation data points with an NRMSE greater than 0.6 are included in the analysis. (a)
Shows evaporation and (b) shows condensation. (c) Only BIN and BULK simulation data
points where the fraction of evaporated mass in one time step is less than 0.25 and (d)
where the fraction of evaporated mass is greater than 0.25 are included in the analysis.

0

BULK100-NU4/BIN100 BULK400-NU4/BIN400 BULK1600-NU4/BIN1600

BULK400-NU2/BIN400 BULK400-NU7/BIN400

-0.4

-0.2

0

In(BULK/BIN Rates)

0.2